PATENT
Attorney Docket No. 202189-0003

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:	)	
	)	
KAWAGUCHI et al.	)	Examiner: Elizabeth A. Bolden
	)	
Application No.: 10/562,268	)	Group Art Unit: 1793
	)	
Filed: December 28, 2005	)	Confirmation No.: 8212
	)	
Title: ALKALI-FREE GLASS	)	

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Alexandria, VA 22314

DECLARATION UNDER 37 C.F.R. § 1.132

Sir:

We, Toru Kawamoto and Noriyuki Yoshida, hereby state as follows:

1. As to Toru Kawamoto:

   (a) I am a citizen of Japan.

   (b) I have a degree from Kyoto University Faculty of Science and Mineralogy and Petrology, awarded March 31, 1985. My academic research was directed to Petrology for Upper Most Mantle and Lower Crustal Rock.

   (c) I have been employed since April 1, 1985 by Nippon Electric Glass Co. Ltd. From June 1, 1985, I was in the Technical Division, Technical Section No. 2 and my research projects and responsibilities included the Development of Glass Composition: Glass for halogen lamp, E-type fiber glass. From August 1, 1990, I was in the Research Division, Melting Research Section and my research projects and responsibilities included the Development of Class Composition: Glass fiber for glass reinforced concrete and

FRP. From October 1, 1997 to present, I have been in the Research and Development Division and my research projects and responsibilities included the Development of Glass Composition: Fiber glass for substrate of electronic component use. In my current position, I also investigate glass fining and research of raw materials for glass batch.

(d) I have carefully studied and understand the specification of the above-named U.S. Patent Application No. 11/562,268.

(e) I have carefully studied and understand the USPTO office communication dated September 17, 2008, in the above-named U.S. Patent Application No. 11/562,268.

(f) I have carefully studied and understand U.S. Patent No. 3,622,296 to *Buehl,* which is cited in the USPTO office communication dated September 17, 2008, in the above-named U.S. Patent Application No. 11/562,268.

2. As to Noriyuki Yoshida:

(a) I am a citizen of Japan.

(b) I have a BA Degree in Molecular Science and Technology from Doshisya University awarded March 1998 and a MA Degree in Chemical Engineering from Doshisya University awarded March 2000.

(c) I have been employed since April 2000 to the present by Nippon Electric Glass Co. Ltd. as a Senior Engineer, Research and Development Division. My research projects and responsibilities include measurement of gas-related properties in glasses and molten glasses and development of glass melting and fining processes.

(d) I have carefully studied and understand the specification of the above-named U.S. Patent Application No. 11/562,268.

(e) I have carefully studied and understand the USPTO office communication dated September 17, 2008, in the above-named U.S. Patent Application No. 11/562,268.

(f) I have carefully studied and understand U.S. Patent No. 3,622,296 to *Buehl,* which is cited in the USPTO office communication dated September 17, 2008, in the above-named U.S. Patent Application No. 11/562,268.

3. We have conducted experiments reproducing the experiments disclosed in U.S. Patent No. 3,622,296 to *Buehl*. The attached Appendix A includes details of these experiments. In this regard, the experiments described in items I, II and III were conducted by Toru Kawamoto and the experiment described in item IV was conducted by Noriyuki Yoshida.

4. Based on our experiments, the experimental data shows the helium content according to the method described in *Buehl* exceeds 2 µl/g. Further, Table 7 shows that to achieve a bubble number of 10/10g (= 100/100g) or less with *Buehl's* conditions, the He-content has to be about 3.9 µl/g and the melt period in He-atmosphere must be longer than about 10 hours.

5. We hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: December 17, 2008 Date: December 17, 2008

Name: Toru Kawamoto Name: Noriyuki Yoshida

Toru Kawamoto Noriyuki Yoshida

Appendix A

I. Introduction to Buehl's Example I (No experiment)

Example I is the only example of Buehl, wherein pyrex glass were used. In each of the cases in Example I, a 30 g batch of the same raw materials of glass with or without a fining agent (1.02% of Cl) was melted at 1450°C in a platinum crucible in an electric oven, in the air-atmosphere for 5-15 hours, or in the air-atmosphere for 1 hour and then in an He-atmosphere for 2.5-15.5 hours. Bubble counting was then conducted for all the samples to compare their fining degrees. The glass compositions and batch compositions are listed in Table 1 and the experiment conditions in Table 2. It is noted that Buehl only describes the batch compositions, so that the glass compositions are calculated based on the compositions of the glass raw materials currently used by Nippon Electric Glass Co., Ltd. (abbreviated to "NEG" hereinafter).

Table 1. Batch compositions and glass compositions calculated therefrom

Glass component (wt%)	Batch A (no fining agent)	Batch B (1.02% of Cl)
SiO_2	80.3	80.3
Al_2O_3	2.15	2.15
B_2O_3	13.24	13.24
Na_2O	4.22	4.67
Cl		1.02
Used reagent (g)		
dried sand	321.8	321.8
anhydrous borax	55.2	49.3
boric acid	26.8	33.9
alumina	8.6	8.6
NaCl		6.8

Table 2. Experiment conditions in Buehl's Example I

Melt temperature	1450℃
Crucible used	Platinum crucible with a diameter of 1.5 inches
Amount of melted batch	30 g
Electric oven used	Unknown
1st melting in air-atmosphere	1 hour
Melting in He-atmosphere	2.5-15.3 hours (2.5 to 6.5 hours for batch with Cl)
He-atmosphere replacement period	30 minutes, 99.8%
He concentration measurement	None
Flow rate of He	Not disclosed
Annealing conditions	Not disclosed

In each case, the glass was taken out from the Pt crucible after being treated in the above conditions, molded into a cube of 2 x 1.5 x 0.2 cm^3 as a sample (density = 2.23 g/cm^3, weight = 1.34 g), and then subjected to bubble counting. The number of bubbles (called "bubble number", hereinafter) in respective samples are converted in the unit of "bubbles/10 g" and listed in Table 3.

Table 3. Bubble numbers (BN) of the samples from Buehl's Example I

Fining agent	Batch	Melt atmosphere	2nd period	BN (/1.34g)	BN (/10g)
None	A	Air (1 hour)	4	7558	56487
			8	1755	13117
			14	403	3012
NaCl	B	Air (1 hour)	4	1450	10837
			8	396	2960
			11	69	516
None	A	Air (1 hour) → He	3	1582	11824
			6.75	23	172
			15.5	5	37
NaCl	B	Air (1 hour) → He	2.5	359	2683
			6.5	5	37

The bubble numbers of all the samples obtained from the batches A & B are shown in Fig. 1 in common logarithms. As shown in Fig. 1, as the same batch A or B was used and the melt period was fixed, the bubble number of the sample staying melted in the He-atmosphere was about 1/100 of that of the sample staying melted in the air-atmosphere. For the batch using Cl as a fining agent and staying melted in He-atmosphere, the longest 2nd melt period was 6.5 hours, with which the bubble number was 37 bubbles/10g (= 370 bubbles/100g), which is much higher than the requirement (< 100 bubbles/100g) for the glass useful in certain optical applications.

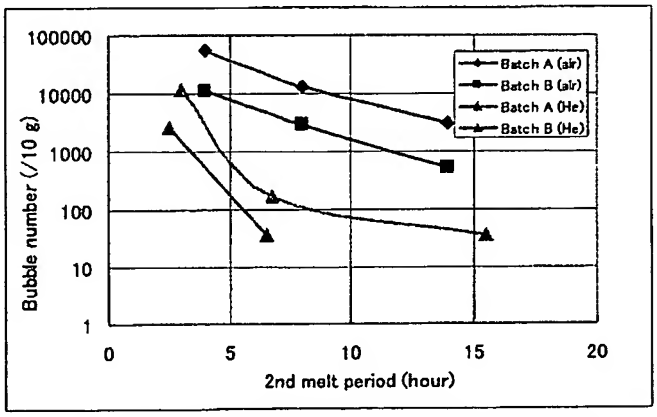

Fig. 1 Bubble numbers of Buehl's samples

II. Experiments of Buehl Example I Reproduced by the applicants

In the Reproduced experiments, the glass composition was obtained according to Buehl's Batch B containing 1.02% of Cl, as shown in Table 4. The silica sand used was Flattery-48M (D_{50} = 195 μm), and the other raw materials were those currently used by the applicants (NEG). As in Buehl's Example I, the melt temperature was set at 1450°C, and each melted batch for producing an He-atmosphere treated sample was firstly placed in the air-atmosphere for 1 hour and then in an He-atmosphere for 2.5 or 6.5 hours. In addition, to obtain small bubble numbers, additional samples with 2nd He-melt periods of 10 hours and 15.5 hours respectively were also made. Samples produced with melting in air-atmosphere were also provided for comparison. The sample glass taken out from the electric oven was molded by carbon plates to be 1 cm thick, rapidly annealed with residual strain, and then observed for the surface. The central portion of the sample is cut in a thickness of about 2 mm, polished and then measured for the bubble number. The He-content of each sample produced with melting in He-atmosphere was measured using a quadrupole mass spectrometer (QMA125, manufactured by Balzers AG Company). For the sample produced with exposure in He-atmosphere for 2.5 hours, because the bubble number is large, a portion thereof at 5 mm from its surface was sampled for measurement. For each of the other samples, a portion thereof at 1 mm from its surface was sampled for measurement.

Table 4. Glass composition in the reproduced experiments and the corresponding batch

Glass component (wt%)	Composition of Batch B
SiO_2	80.3
Al_2O_3	2.15
B_2O_3	13.24
Na_2O	4.67
Cl	1.02
Raw materials (g) used in reproduced experiments, blended for 20 minutes	Batch used in reproduced experiments (by NEG)
Flattery-48M silica sand	22.96
anhydrous borax	3.52
boric acid	2.42
alumina (A-21 GL)	0.61
calcined pure salt	0.49
Total (g)	30

Table 5. Conditions of reproduced experiments (compared with Buehl)

	Reproduced experiments	Buehl's Example I
Melt temperature	1450°C	
Crucible used	Triangular Pt/Au crucible of 100 ml	Pt crucible with a diameter of 1.5 inches
Amount of melted batch	30 g	
Electric oven	O_2-atmosphere electric oven (FUB722PA made by Advantec Toyo Company)	Unknown
1st melting in air-atmosphere	1hour	
Melting in He-atmosphere	2.5, 6.5, 10 or 15.5 hours	2.5-15.3 hours (2.5 or 6.5 hours for batch with Cl)
He-atmosphere replacement period	30 minutes/98% → 60 minutes/99.8% (then fixed)	30 minutes/99.8%
He concentration measurement	QMA125	None
Flow rate of He	5 l/min (initial 30 minutes) → 3 l/min	unknown
Annealing conditions	Pressed to about 10 mm thick after melting → rapid annealing	unknown

III. Results of Reproduced Experiments

1. Bubble Number:
Referring to Table 6 and Fig. 2, the sample obtained with melting in He-atmosphere for 2.5 hours had a bubble number of 820/10g, which was about 1/3 of that (2612/10g) of the sample obtained with melting in the air-atmosphere. When the 2nd meld period was 6.5 hours, the He-atmosphere treated sample had a bubble number of 33/10g, which was about 3/100 of that (1127/10g) of the corresponding air-atmosphere treated sample. As the 2nd meld period was up to 10 or 15.5 hours, the He-atmosphere treated sample had a bubble number of 1/10g and became well fined, while the corresponding air-treated samples had bubble numbers of 680/10g and 368/10g, respectively.

2. He-content:
For He-atmosphere treated samples, when the 2nd melt period was 6.5 hours, the He-content of the glass was 2.291 µl/g, which was higher than the upper limit (2.0 µl/g) of the range of claim 1. When the 2nd melt period was increased to 10 hours,

the He-content was increased to 3.955 µl/g. When the 2nd melt period was increased to 15.5 hours, the He-content was 3.396 µl/g. See Table 6 and Fig. 3. Thus, the He-content reached equilibrium after about 10 hours.

3. Comparison:
As compared with the best mode of Buehl's Example I, i.e., a bubble number of 37/10g for the sample obtained with melting in the He-atmosphere for 6.5 hours, the result (33/10g) of the corresponding reproduced experiment made by the applicants was very close thereto. See Table 6 and Fig. 2. This demonstrates that the samples made by the applicants according to Buehl's disclosure were substantially the same as those made by Buehl.

Table 6: Experiment data of the reproduced experiments (compared with Buehl)

	Data of the reproduced experiments by the applicants			Buehl
2nd melt period (hr)	Air-atmosphere BN (/10g)	He-atmosphere BN (/10g)	He-content (µl/g)	He-atmosphere BN (/10g)
2.5	2612	820	0.951	2683
6.5	1127	33	2.291	37
10	680	1	3.955	-
15.5	368	1	3.396	-

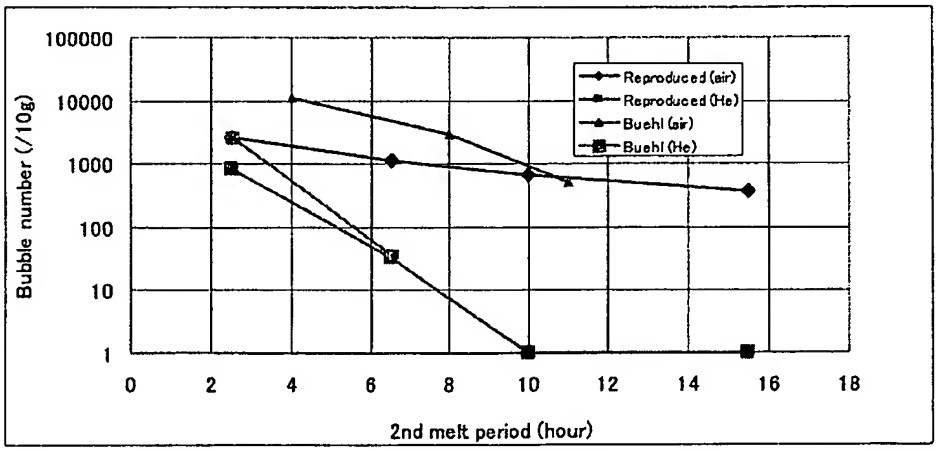

Fig. 2 Bubble numbers in the reproduced experiments (compared with Buehl's Batch B having 1.02% of Cl)

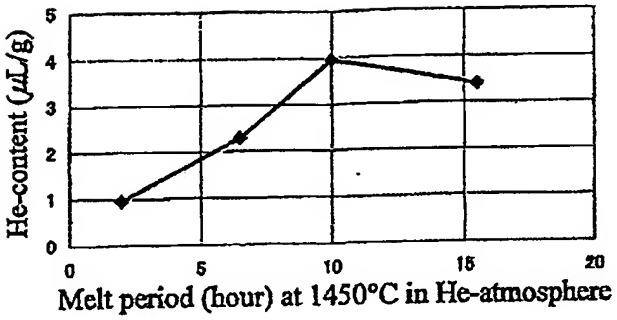

Fig. 3: Relationship between the He-content in the glass and the melt period in the He-atmosphere in the reproduced experiments

The He-contents and the bubble numbers of the samples treated in He-atmosphere for different melt periods in the applicants' reproduced experiments for Buehl's Example I are put together in Table 7 to show the relationship between any two of He-content, the bubble number and the melt period in the He-atmosphere.

Table 7

	Reproduced experiments for Buehl		
Glass material	Pyrex		
Fining agent	1.02% of Cl		
Melt period (hr) in He-atmosphere	6.5	10	15.5
He-content (μl/g)	2.291	3.955	3.396
Bubble number (/10g)	33	1	1

Table 7 shows that to achieve a bubble number of 10/10g (= 100/100g) or less with Buehl's conditions, the He-content has to be about 2.5 μl/g and the melt period in He-atmosphere must be longer than about 7.5 hours.

IV. More Disadvantages for a He-content Greater than 2 μl/g

Noble gases like helium and neon are classified as physical dissolution gases, of which the solubility is reported to increase with an increase in the temperature, as shown in Fig. 4 (from James E. Shelby, "Introduction to Glass Science & Technology, 2nd Edition", in the Royal Society of Chemistry, p109 (2005), which shows the temperature-dependent solubility of He in borosilicate glass (BSG).

It is noted that the glass used in *Buehl's* Example I is a type of BSG. As shown in Fig. 4, when the melt temperature in He-atmosphere is higher than 1400°C, the He-

content in the BSG is larger than 2 μl/g and increases along with the melt temperature. The temperature of 1450°C in *Buehl* is annotated on FIG. 4.

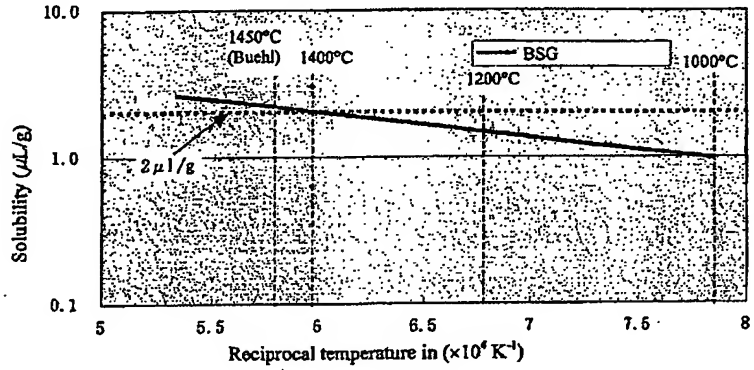

Fig. 4

According to Fig. 4, when a BSG melt previously saturated with helium and/or neon at a high temperature (e.g., 1450°C) is rapidly cooled as in *Buehl*, the gas in the BSG is in an over-saturated state. Hence, as the BSG is related to *a temperature within a range of about 1000-1200°C required for glass molding* to be molded into a glass product, the excess He/Ne forms bubbles in the glass - a phenomenon called reboiling. Since a glass melt has a relatively high viscosity at a temperature of about 1000-1200°C, the bubbles do not ascend and disappear easily so that significant defects are seen in the glass product fabricated.